Qubit coherence control in a nuclear spin bath

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Coherent dynamics of localized spins in semiconductors is limited by spectral diffusion arising from dipolar fluctuation of lattice nuclear spins. Here we extend the semiclassical theory of spectral diffusion for nuclear spins $I = 1/2$ to the high nuclear spins relevant to the III-V materials and show that applying successive qubit π-rotations at a rate approximately proportional to the nuclear spin quantum number squared (I^2) provides an efficient method for coherence enhancement. Hence robust coherent manipulation in the large spin environments characteristic of the III-V compounds is possible without resorting to nuclear spin polarization, provided that the π-pulses can be generated at intervals scaling as I^{-2}.

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I. INTRODUCTION

The ability to enhance coherence times of localized electron or nuclear spin qubits in a semiconductor environment is central for the development of a spin based solid state quantum computer[1]. Qubit coherence in these devices appears to be limited by the interaction with nuclear spins composing the material[2,3], with other mechanisms being less important. A detailed comparison of the many mechanisms contributing to localized spin decoherence in semiconductors is given in Refs. 3,4. Experimental evidence of the relative contributions of different decoherence channels is given for silicon in Refs. 5,6. Here we consider qubit coherence under the strong effective magnetic fields and low temperatures required for single spin read-out ($B > 5$ Tesla and $T \sim 100$ mK, see Ref. 7). Under these conditions, environmental nuclear spin fluctuations result primarily from inter-nuclear dipolar coupling, which becomes a source for time-dependent noise in the qubit Zeeman energy and for its consequent decoherence. This channel for phase relaxation is usually denoted spectral diffusion[4,5,8,9].

It was pointed out recently that substantial nuclear polarization can suppress spectral diffusion[2,4]. However, the requirement for nuclear polarization adds significant device overhead, since polarizations on the order of 95% or more are needed in order to achieve significant coherence enhancement (See Fig. 3 of Ref. 10). Recently, several schemes for nuclear polarization were proposed[11], but it is still not clear whether these will be effective enough to compete with the nuclear spin diffusion rate [$1 - 10$ KHz, see Eq. (12) below].

Several pulse sequences have been designed to average out the effect of nuclear-nuclear dipolar coupling, most notably WAHUHA and Lee-Goldburg schemes[12]. Because these sequences are based on nuclear spin excitation, they inevitably generate a drastic time-dependent drift in the spin qubit Zeeman frequency. The amplitude of this drift can be as much as 50 G (typical hyperfine line-width for a localized electron in a III-V compound), fluctuating at the nuclear spin rotation frequency (typically $10^4 - 10^6$ Hz depending on the rf field intensity). The effect of these pulses is additional field fluctuation which adds to the decoherence rate and makes qubit control impossible. Under such a protocol, the fidelity of quantum operations would be drastically affected in a negative sense. Conversely, the nuclear spins causing spectral diffusion are subject to an inhomogeneous field arising from the interaction with the qubit. This leads to a wide spectrum of nuclear resonance frequencies, requiring broad-band excitation in order to perform nuclear spin rotation. Broad-band excitation usually requires high power deposition, which is incompatible with the MilliKelvin temperatures needed for single spin measurement[13]. These arguments suggest that *any scheme for spectral diffusion control must rely on excitation of the qubits themselves, and not excitation of the nuclei.*

Here we consider the Carr-Purcell-Meiboom-Gill sequence (CPMG)[14,15] as a scheme for spectral diffusion control. The CPMG pulse sequence consists of the successive application of qubit π-rotations perpendicular to $\boldsymbol{B}$ at time instants $(2n+1)\tau$, $n = 0, 1, 2, \ldots$. This procedure leads to the formation of spin echoes at instants $(2n + 2)\tau$, whose coherence is substantially enhanced when compared to qubit evolution in the absence of π-pulsing. Coherence enhancement takes place provided τ remains below a threshold τ_c, the typical time scale for environmental fluctuations. As τ is decreased further, the coherence gain can be substantially amplified, with an increased cost in the number of π pulses. Here we develop a microscopic theory which is able to predict, without any fitting parameters, the inter-pulse time τ_n needed to provide n times more coherence for a spin qubit subject to nuclear-induced spectral diffusion in a bath of general spin I. Our explicit calculations reveal that in realistic devices nuclear spin noise can only be suppressed if τ is inversely proportional to the nuclear spin quantum number I squared, establishing the overhead requirement for CPMG control of an isolated spin coupled to a nuclear bath of general spin I.

Available spectral diffusion theories have focused solely

on the case of $I = 1/2$ environments, using two-level telegraph noise models to analyze the coherence time of a central spin-1/2[4,8]. However many solid state spin-based proposals rely on high spin environments, an important example being the GaAs ($I = 3/2$) quantum dot. Here one often has to add In ($I = 9/2$) to enable single spin read-out[16,17] in addition to a substantial amount of Al ($I = 5/2$) to achieve confinement[18]. InAs self assembled quantum dots are also promising with regard to optical manipulation and transport[11,16,19,20]. The requirements for CPMG control in these structures are not yet known. In this work we first characterize spectral diffusion for $I > 1/2$ environments and then analyse coherence control with CPMG. Our results establish CPMG as a remedy for the absence of $I = 0$ isotopes in III-V materials[6] as long as existing schemes for fast and precise spin rotation are further developed[18,19,21].

II. SPECTRAL DIFFUSION DECAY INDUCED BY $I > 1/2$ NUCLEAR SPINS

Our qubit/spin I bath Hamiltonian is given by

$$\begin{aligned}\mathcal{H} &= \gamma_S B S_z - \gamma_I B \sum_n I_{nz} \\ &\quad + \sum_n A_n I_{nz} S_z - 4 \sum_{n<m} b_{nm} I_{nz} I_{mz} \\ &\quad + \sum_{n<m} b_{nm}(I_{n+}I_{m-} + I_{n-}I_{m+}). \qquad (1)\end{aligned}$$

The first term describes the qubit two-level structure, γ_S and S_z being its gyromagnetic ratio and spin operator. The nuclear spin operators $\boldsymbol{I}_n$ are coupled to the qubit through the parameters A_n. These are determined by hyperfine coupling (for an electron spin qubit) or by dipolar interaction (for a nuclear spin qubit). The inter-nuclear dipolar coupling b_{nm} is responsible for nuclear spin fluctuations (for details see Ref. 4). In particular, the last term of Eq. (1) induces "flip-flop" transitions between pairs of nuclear spins. When such a transition takes place, the qubit level spacing shifts by $2\Delta_{nm} = |A_n - A_m|$, leading to phase randomization and to decoherence. The dipolar coupled spin-I lattice acts as a bath which is coupled to the qubit with strength $\Delta_{nm} \equiv \Delta$.

In the semiclassical approach to spectral diffusion[4], the intricate nuclear spin evolution stemming from Eq. (1) is effectively described by a set of uncorrelated classical stochastic transitions (flip-flops), with a characteristic rate Γ obtained from first principles. It is appropriate to use a sudden-jump model for the description of these processes because the time scale for thermal fluctuation is much shorter than $\frac{1}{\Gamma}$ ($\hbar/k_B T \ll$ ns$\ll \frac{1}{\Gamma} \sim$ ms). Correlation between flip-flop events of fluctuating pairs located near each other become important only when these pairs flip-flop several times within τ (or equivalently $\Gamma\tau \gg 1$). For relevant time scales ($2\tau < T_2$) we can justify our neglect of flip-flop correlation *a posteriori* by noting that the condition $\mathrm{Max}\,\Gamma\tau \gg 1$ is never realized in the physical cases considered here.

A. Calculation of the flip-flop transition rates in a spin-I bath

In the semiclassical theory, nuclear spin fluctuation is decoupled from the qubit by setting $S_z \to 1/2$ in the full Hamiltonian Eq. (1). This leads to an effective Hamiltonian $\mathcal{H}'$ for nuclear spin evolution under the inhomogeneous field produced by the qubit,

$$\mathcal{H}' = \mathcal{H}_0 + \sum_{n<m} F_{nm}(t), \qquad (2)$$

$$\mathcal{H}_0 = -\sum_n \left(\gamma_I B - \frac{1}{2} A_n\right) I_{nz} - 4 \sum_{n<m} b_{nm} I_{nz} I_{mz}, \quad (3)$$

$$F_{nm}(t) = b_{nm}\left(I_{n+}I_{m-}\mathrm{e}^{-i\omega t} + I_{n-}I_{m+}\mathrm{e}^{i\omega t}\right). \quad (4)$$

In Eq. (4), we have introduced a fictitious frequency ω which allows a connection to the method of moments[4]. After we calculate the flip-flop rate, we will take the limit $\omega \to 0$.

Without loss of generality, we assume $n = 1$, $m = 2$. The eigenstates of $\mathcal{H}_0$ are given by $|M_1, M_2, \ldots\rangle$, with $M_i = -I, -I+1, \ldots, I$. The flip-flop operators $F_{12}(t)$ do not connect all states because they conserve $\mathcal{J} = (M_1 + M_2)/2$. Hence flip-flop dynamics break the pair Hilbert space into disconnected subspaces labeled by $\mathcal{J} = I, I-1/2, \ldots, -I$. Each subspace has dimension $2(I - |\mathcal{J}|) + 1$ and can thus be alternatively labeled by a pseudo-spin $I' = I - |\mathcal{J}|$, which is seen to correspond to projection of the pseudo-spin operator $(\boldsymbol{I}_1 - \boldsymbol{I}_2)/2$ with projection index $M' = (M_1 - M_2)/2 = -I', \ldots, I'$. Each nuclear pair contains two subspaces for each $I' = 0, 1/2, 1, \ldots, I - 1/2$ (since $\mathcal{J}$ can be positive or negative) and one subspace with $I' = I$.

The Fermi Golden rule rate for a transition $M' \leftrightarrow M' - 1$ between the state $|M'\rangle = |M_1, M_2, M_3, \ldots, M_N\rangle$ and $|M' - 1\rangle = |M_1 - 1, M_2 + 1, M_3, \ldots, M_N\rangle$ is given by

$$\begin{aligned}\Gamma &= 2\pi b_{12}^2 \sum_{M_3,\ldots,M_N} p(M_3, \ldots, M_N) \\ &\quad \times \left|\langle M' - 1|I_{1-}I_{2+}|M'\rangle\right|^2 \delta(E^0_{M'-1} - E^0_{M'} - \omega), (5)\end{aligned}$$

where $p(M_3, \ldots)$ are Boltzmann probabilities for the unperturbed energies $E^0_{M'}$. It is convenient to define an auxiliary function

$$\rho(\omega) = \sum_{M_3,\ldots,M_N} p(M_3, \ldots, M_N)\delta(E^0_{M'-1} - E^0_{M'} - \omega), \qquad (6)$$

which can be interpreted as a density of states. After evaluating the matrix element in Eq. (5) we get

$$\begin{aligned}\Gamma &= 2\pi b_{12}^2(2I-I'+M')(2I-I'-M'+1)\\ &\quad\times(I'+M')(I'-M'+1)\rho(\omega), \qquad (7)\end{aligned}$$

where we recall that $I' = I - |M_1+M_2|/2$ and $M' = (M_1-M_2)/2$. We can now calculate explicitly the moments of the function $\rho(\omega)$. For example, $\int \rho(\omega)d\omega = 1$, while the first moment is given by

$$\begin{aligned}\overline{\omega}_{12} &= \int_{-\infty}^{\infty}\omega\rho(\omega)d\omega\\ &= \sum_{M_3,\ldots,M_N} p(M_3,\ldots,M_N)(E^0_{M'-1}-E^0_{M'})\\ &= \frac{1}{2}(A_2-A_1) - 4b_{12}(M_1-M_2-1)\\ &\quad+4\sum_{k\neq 1,2}(b_{1k}-b_{2k})\langle m_k\rangle. \qquad (8)\end{aligned}$$

Here the thermal average $\langle m_k\rangle = \partial \ln Z/\partial\beta'$ can be calculated from the partition function

$$Z = \sum_{m=-I,\ldots,I} \mathrm{e}^{\beta' m}, \qquad (9)$$

with $\beta' = \hbar\gamma_I B/k_B T$ (for $B \gg I\sum_n b_{nm}/\gamma_I \sim 10$ G we can neglect the dipolar energy in the Boltzmann distributions). The second moment is given by

$$\begin{aligned}\kappa_{12}^2 &= \int(\omega-\overline{\omega}_{12})^2\rho(\omega)d\omega\\ &= 16\sum_{k\neq 1,2}(b_{1k}-b_{2k})^2\langle(m_k-\langle m_k\rangle)^2\rangle, \qquad (10)\end{aligned}$$

where $\langle(m_k-\langle m_k\rangle)^2\rangle = \partial^2\ln Z/\partial^2\beta'$. The fourth moment q_{12}^4 can also be calculated, allowing us to show that the ratio $q_{12}^4/(3\kappa_{12}^2) \sim 1$. Thus the Gaussian function provides a reasonable fit to Eq. (6),

$$\rho(\omega) \approx \frac{1}{\sqrt{2\pi\kappa_{12}^2}}\exp\left[-\frac{(\omega-\overline{\omega}_{12})^2}{2\kappa_{12}^2}\right]. \qquad (11)$$

The final expression for the flip-flop rate for a nuclear spin pair (n,m) is[22]

$$\begin{aligned}\Gamma_{M'} &= (2I-I'+M')(2I-I'-M'+1)(I'+M')\\ &\quad\times(I'-M'+1)\frac{\sqrt{2\pi}b_{nm}^2}{\kappa_{nm}}\exp\left(-\frac{\omega_{nm}^2}{2\kappa_{nm}^2}\right). \qquad (12)\end{aligned}$$

When $\beta' \ll 1$, there is only a weak temperature and B field dependence in Eq. (12). For example, $T = 100$ mK and $B = 10$ T leads to $\beta' \sim 0.1$. In this approximation we have $\langle m_k\rangle \approx 0$ and $\langle(m_k-\langle m_k\rangle)^2 \approx I(I+1)/3$. This leads to the approximate expressions

$$\overline{\omega}_{nm} \approx \Delta_{nm} - 4b_{nm}(2M'-1), \qquad (13)$$

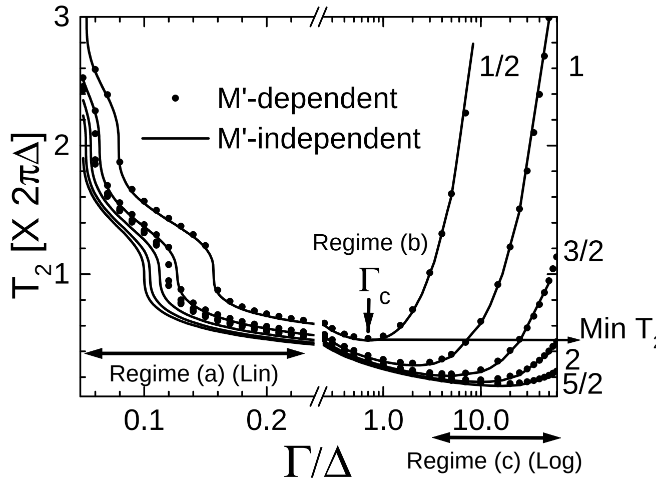

FIG. 1: Behavior of $1/e$ decay times for the first CPMG echo in pseudo-spin subspaces $I' \leq 5/2$ as a function of flip-flop rate Γ. Circles: calculation with M'-dependent rates $\Gamma_{M'}$. Solid lines: calculation with average rate $\langle\Gamma_{M'}\rangle$. Each curve achieves minimum coherence at $\Gamma = \Gamma_c$, which is identified as a critical correlation rate. Small Γ (or strong coupling Δ) imply low frequency noise with time correlation [regime(a)], while large Γ (or weak coupling Δ) imply motional narrowing or white noise behavior [regime (c)]. Note the strong I' dependence in the weak coupling region. Here we have $\mathrm{Min}\,(T_2) \approx 6/\Delta(2I'+1)$, and $\Gamma_c \approx (2I'+1)^3/12$.

$$\kappa_{nm}^2 \approx \frac{16}{3}I(I+1)\sum_{i\neq n,m}(b_{ni}-b_{mi})^2. \qquad (14)$$

κ_{nm} represents the amount of energy the dipolar system can supply for a nuclear flip-flop to take place. Nuclear spins located near the center of the electron wave function are characterized by $\overline{\omega}_{nm} \gg \kappa_{nm}$, with exponentially small flip-flop rates. These are said to form a frozen core which does not contribute to spectral diffusion (a similar effect has been reported in optical experiments, see Ref. 23). On the other hand nuclei satisfying $\overline{\omega}_{nm} \sim \kappa_{nm}$ possess appreciable flip-flop rates together with a sizable hyperfine shift Δ_{nm}. These are located in a shell around the center of the electronic wave function, and are responsible for most of the spectral diffusion coherence decay[3,4]. Below we show that to a very good approximation the M' dependence in Eq. (12) averages out, so that we may consider just the average rate

$$\Gamma(I',I) = \langle\Gamma_{M'}\rangle \approx \xi(I',I)\,\Gamma(I,I) \sim II'^2, \qquad (15)$$

with the parameter $\xi(I',I)$ connecting different subspaces of a spin-I pair,

$$\xi(I',I) = \frac{I'(I'+1)\,[1+5I(2I+1)+I'(2I'-10I-3)]}{I(I+1)\,[1+2I(I+1)]}. \qquad (16)$$

B. Random walk theory for the fluctuation of a I' subspace

We now formulate a random walk theory for the coupling of the qubit to the flip-flop dynamics in each pseudo-spin subspace I' and then construct the coherence decay as a weighted average over all pseudo-spin subspaces. Our model assumes that each I' subspace fluctuates independently, i.e. pairs are uncorrelated. As a result, the qubit Zeeman frequency is subjected to several random fields of the form $\Delta\sigma_{I'}(t)$, where $\sigma_{I'}$ is a hard wall random walk variable assuming the values

$$\sigma_{I'}(t) = \{-2I', -2(I'-1), \ldots, 2I'\}\,. \tag{17}$$

The theory for qubit decoherence resulting from the two $I' = 1/2$ subspaces is described in Ref. 4. It has a simple structure because for $I' = 1/2$, $\sigma_{I'}(t)$ can be written as $(-1)^{N(t)}$, with $N(t)$ a Poisson random variable with parameter Γt. The generalization of this telegraph process to the multi-level noise present when $I' > 1/2$ is achieved by defining an occupation probability vector $\boldsymbol{p}(t)$ and its Markovian evolution operator $U(t)$:

$$\begin{aligned}\boldsymbol{p}(t) &= (p_{-I'}, p_{-I'+1}, \ldots, p_{I'}), &(18)\\ &= \exp\left(-t\boldsymbol{A}\right)\cdot\boldsymbol{p}(0) &(19)\\ &\equiv \boldsymbol{U}(t)\cdot\boldsymbol{p}(0). &(20)\end{aligned}$$

For first-order flip-flop transitions we have

$$\dot{p}_i = \Gamma_i\, p_{i-1} - (\Gamma_i + \Gamma_{i+1})p_i + \Gamma_{i+1}\, p_{i+1}, \tag{21}$$

$$A_{ij} = -\Gamma_i\delta_{i-1,j} + (\Gamma_i + \Gamma_{i+1})\delta_{i,j} - \Gamma_{i+1}\delta_{i+1,j}. \tag{22}$$

We can determine the echo decay for all Markovian processes described by $\boldsymbol{A}$ using the time-ordered correlation function ($t_k \geq t_{k-1} \geq \cdots \geq t_1$)

$$\begin{aligned}\langle\sigma_{I'}(t_k)\cdots\sigma_{I'}(t_1)\rangle &= (1\cdots1)\cdot\boldsymbol{\Sigma}_{I'}\cdot\boldsymbol{U}(t_k - t_{k-1})\\ \cdot\boldsymbol{\Sigma}_{I'}\cdot\boldsymbol{U}(t_{k-1}-t_{k-2})&\cdots\boldsymbol{\Sigma}_{\boldsymbol{I'}}\cdot\boldsymbol{U}(t_1)\cdot\boldsymbol{p}(0), \qquad (23)\end{aligned}$$

defined by correlations between values of the spin state matrix

$$\boldsymbol{\Sigma}_{I'} = \mathrm{diag}\left\{-2I', -2(I'-1), \ldots, 2I'\right\} \tag{24}$$

at times $t_1, t_2, \ldots, t_k$. The Hahn echo is then calculated using a double average

$$v_{I'}(2\tau) = \left\langle\left\langle\exp\left(i\int_0^{2\tau} s(t)\sigma_{I'}(t)dt\right)\right\rangle_{\sigma_{I'}(0)=\sigma}\right\rangle_{\sigma}, \tag{25}$$

where the echo function is $s(t) = 1$ for $t \leq \tau$ and $s(t) = -1$ for $t > \tau$ (we set $\Delta = 1$ for simplicity, since it will be recovered later as the unit of time). The inner average is over $\sigma_{I'}(t)$ trajectories starting from $\sigma_{I'}(0) = \sigma$. The outer average is a thermal average over all possible initial states σ. The n-th echo of the CPMG sequence can then be simply obtained from the n-th power of Eq. (25)[15,24].

The difficulty of evaluating Eq. (25) lies in ensuring a proper treatment of $s(t)$. We propose here a matrix method that enables the incorporation of any step-wise constant $s(t)$ explicitly. After expanding and rearranging the integrals we obtain

$$\begin{aligned}v_{I'}(2\tau) &= \sum_{\sigma} p_\sigma(0)\sum_{j,k=0}^{\infty}(-i)^j(i)^k\int_\tau^{2\tau}dt'_j\ldots\int_\tau^{t'_2}dt'_1\\ &\int_0^\tau dt_k\ldots\int_0^{t_2}dt_1\left\langle\left[\sigma(t'_j)\cdots\sigma(t'_1)\right]\right.\\ &\left.\times\left[\sigma(t_k)\cdots\sigma(t_1)\right]\right\rangle_\sigma, \qquad (26)\end{aligned}$$

where $\tau \geq t_k \geq \ldots \geq t_1 \geq 0$ and $2\tau \geq t'_j \geq \ldots \geq t'_1 \geq \tau$ are partitions before and after the π pulse. Using Eq. (23) and the Markovian identity $\boldsymbol{U}(t'_1 - t_k) = \boldsymbol{U}(t'_1-\tau)\cdot\boldsymbol{U}(\tau - t_k)$, Eq. (26) can be written as

$$v_{I'}(2\tau) = \frac{1}{2I'+1}(1,\ldots,1)\cdot\boldsymbol{M}_-\cdot\boldsymbol{M}_+\cdot\begin{pmatrix}1\\ \vdots\\ 1\end{pmatrix}, \tag{27}$$

where the initial probabilities are simply $p_\sigma(0) = 1/(2I'+1)$. The matrices $\boldsymbol{M}_\pm$ are given by (after the substitution $t''_j = t'_j - \tau$)

$$\boldsymbol{M}_-(\tau) = \sum_{j=0}^{\infty}(-i)^j\int_0^\tau dt''_j\boldsymbol{m}_j(t''_j), \tag{28}$$

$$\boldsymbol{M}_+(\tau) = \sum_{k=0}^{\infty}(i)^k\int_0^\tau dt_k\boldsymbol{U}(\tau - t_k)\cdot\boldsymbol{m}_k(t_k), \tag{29}$$

where $\boldsymbol{m}_l(t_l)$ is obtained from the recurrence relation

$$\boldsymbol{m}_l(t_l) = \int_0^{t_l}dt_{l-1}\boldsymbol{\Sigma}_{I'}\cdot\boldsymbol{U}(t_l - t_{l-1})\cdot\boldsymbol{m}_{l-1}(t_{l-1}), \tag{30}$$

$$\boldsymbol{m}_1(t_1) = \boldsymbol{\Sigma}_{I'}\cdot\boldsymbol{U}(t_1). \tag{31}$$

Going to Laplace space the convolution integrals are simply converted to multiplications,

$$\widetilde{\boldsymbol{m}}_l(r) = \int_0^\infty \mathrm{e}^{-rt}\boldsymbol{m}_l(t)dt = \left[\boldsymbol{\Sigma}_{I'}\cdot\widetilde{\boldsymbol{U}}(r)\right]^l. \tag{32}$$

Using Eqs. (28), (29) we obtain closed expressions for the Laplace transforms of the matrices $\boldsymbol{M}_\pm$,

$$\widetilde{\boldsymbol{M}}_-(r) = \frac{1}{r}\frac{\boldsymbol{1}}{\boldsymbol{1} + i\boldsymbol{\Sigma}_{I'}\cdot\widetilde{\boldsymbol{U}}(r)}, \tag{33}$$

$$\widetilde{\boldsymbol{M}}_+(r) = \widetilde{\boldsymbol{U}}(r)\frac{\boldsymbol{1}}{\boldsymbol{1} - i\boldsymbol{\Sigma}_{I'}\cdot\widetilde{\boldsymbol{U}}(r)}. \tag{34}$$

These general expressions for multi-level Markovian echo can be further simplified when written as a function of the eigenvectors and eigenvalues of $\boldsymbol{A}$.

Solving Eqs. (27), (33)-(34), using (20)-(22), allows evaluation of the decoherence time T_2 from the $1/e$ decay of the Hahn echo, Eq. (27). An analytical expression for the Hahn echo is obtained using *Mathematica*[25]. Fig. 1 shows T_2 as a function of average flip-flop rate Γ for several I' subspaces. First we note that for all I', averaging over the M' dependences in $\Gamma_{M'}$ is an excellent approximation [resulting from the outer average in Eq. (25)]. For each I', we identify a critical bath correlation rate Γ_c for which the minimum decay time T_2 is found. This leads to a natural division into three different regimes which can be seen to possess different capabilities for coherence enhancement by CPMG sequences. We recall that application of a CPMG sequence to Hahn echo given by $\exp[-(2\tau/T_2)^d]$ transforms this to $\exp(-2n\tau/T_{\rm CPMG})$ where $T_{\rm CPMG} = [T_2/(2\tau)]^{d-1}T_2$ provides a measurement of the coherence enhancement which is obtained for all $d > 1$ (no enhancement results for $d = 1$)[15].

We refer to Fig. 1 for an illustration of regimes (a), (b), (c). Regime (a) holds for $\Gamma/\Delta \ll \Gamma_c$. This is the regime of sudden-jump spectral diffusion (strong coupling Δ) which has oscillations in the echo envelope due to precession under a finite Δ. Assuming that at most two flip-flops take place, we obtain an expansion up to second order in (Γ/Δ),

$$
\begin{aligned}
v_{I'}(2\tau) \quad &\approx \frac{2\mathrm{e}^{-\rho}}{2I'+1}\left\{1+\frac{2I'-1}{2}\mathrm{e}^{-\rho}+\left(\frac{\Gamma}{\Delta}\right)\sin\delta\right. \\
&\times\left[1+(2I'-1)\mathrm{e}^{-\rho}\right]+\left(\frac{\Gamma}{\Delta}\right)^2\left[1-\cos\delta\right. \\
&\left.\left.+\frac{2I'-1}{2}\mathrm{e}^{-\rho}\left(1-\cos 2\delta\right)\right]\right\}, \qquad (35)
\end{aligned}
$$

where $\rho = 2\tau\Gamma$ and $\delta = 2\tau\Delta$. When $\tau < 1/\Delta$, CPMG provides coherence enhancement proportional to $(T_2/\tau)^2$. Regime (b) holds for $\Gamma/\Delta \sim \Gamma_c$. This is the regime of critical spectral diffusion, which occurs when T_2 reaches a minimum. The minimum is a result of the inevitable mixing of the random walk noise in a finite walk, such as that in the pseudo-spin subspace considered here. Coherence enhancement is approximately proportional to (T_2/τ) for $\tau < 1/\Delta$. Finally, regime (c) satisfies $\Gamma/\Delta \gg \Gamma_c$. This corresponds to the continuum or weak coupling regime. For $2\tau \sim T_2$ nuclear spin fluctuation is so fast that the coherence of the central spin-1/2 is enhanced. Motional narrowing takes place when the stochastic process traverses the random walk space several times; hence it is insensitive to all eigenvalues of $\boldsymbol{A}$ except for the lowest non-zero eigenvalue a^* and associated eigenvector $\boldsymbol{v^*}$. The latter observation allows us to derive an analytical expression for the decoherence time at motional narrowing, when qubit spin echo $v_{I'} \approx \exp(-2\tau/T_2)$ with

$$
T_2 \approx \frac{2I'+1}{2I'}\frac{a^*}{v_1^*\sum v_j^*(\boldsymbol{\Sigma}_{I'})_{jj}}\frac{\Gamma}{\Delta^2} \propto I'^{-4}. \qquad (36)
$$

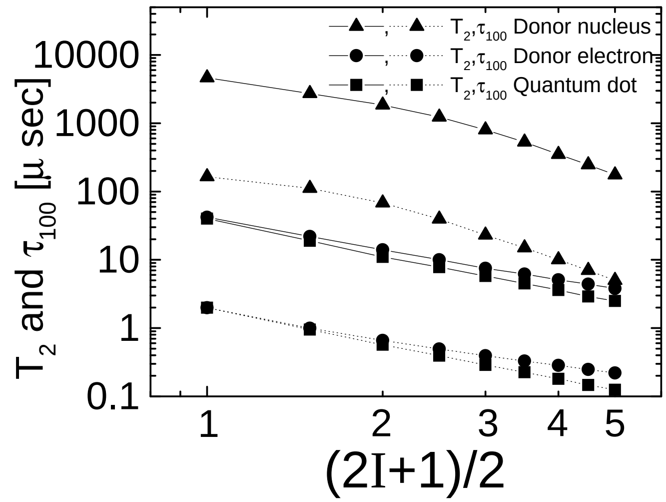

FIG. 2: Coherence times T_2 and inter-pulse times τ_{100} required for 100× coherence enhancement as a function of nuclear spin quantum number I. We find that both T_2 and τ_{100} scale as $\sim 1/I^{1.5}$.

When Eq. (36) holds, no CPMG enhancement is observed. We note however that as τ decreases below a certain threshold value $\tau_c \approx 1/(a^*\Gamma) \approx (2I'+1)^2/(8\Gamma)$, crossover to $v_{I'} \sim \exp(-\tau^3)$ behavior takes place and consequently the CPMG pulse sequence will again provide effective enhancement of coherence. The threshold value τ_c is related to continuous Gaussian noise, since the limit $\Gamma \to \infty$, $\Delta \to 0$ and $I' \to \infty$ with τ_c and $I'\Delta$ constant is given by Eq. (A11) of Ref. 9.

In order to determine the dependence of spin coherence on the bath nuclear spin value I, we make an average of Eq. (27) over all I' subspaces and nuclear species s. We recall that most materials considered here are a mixture of different isotopes. In a strong external field, only flip-flop between the same nuclear species needs to be considered, since flip-flop between different species is strongly suppressed due to an offset in hyperfine shifts. The resulting echo envelope is

$$
\langle v\rangle_I(2\tau) = \sum_{s,I'} p(s,I')v_{I'}[2\tau,\xi(I',I)\Gamma], \qquad (37)
$$

where $p(s,I') \propto f_s^2(2I'+1)\cosh[2(I-I')\hbar\gamma_I B/k_B T]$ with f_s the isotopic abundance for nuclear species s. Eq. (37) shows that substantial nuclear spin polarization (or extremely low nuclear spin temperature T) would be required to suppress spectral diffusion. For example, $p(0) = 0.9$ (corresponding to 90% of nuclear spins having maximum magnetization I) increases T_2 by a factor of 2 only, while for the completely unpolarized case, $p(I') = 2(2I'+1)/(2I+1)^2$.[10] Numerical evaluation shows that the dependence of Eq. (37) on Γ/Δ is qualitatively similar to that seen for the individual I' subspaces in Fig. 1, but with different scaling behavior for the characteristic quantities. For example, $\Gamma_c \approx [(2I+1)/3]^{2.7}$, allowing possible crossover between the three regimes as

I increases.

III. NUMERICAL RESULTS

The complete coherence decay is obtained as a product of decays from multiple fluctuator pairs n, m. Most $I > 1/2$ materials of interest have similar dipolar couplings (b_{nm}) so that we may conclude that CPMG control depends primarily on the shape of the qubit wave function (which determines the distribution of Δ_{nm}) and on the magnitude of the nuclear spin I. Fig. 2 shows our numerical results for T_2 and τ_{100} (inter-pulse time needed for $100\times$ coherence enhancement) for the case of donor impurities (electron and nuclear spin qubit) and a III-V quantum dot (electron spin qubit – see Ref. 7). For an electron spin it turns out most nuclear pairs are fluctuating in regime (a) with $\Gamma T_2 \ll 1$, implying the threshold for coherence enhancement τ_c is quite close to T_2. As τ decreases below $\tau_c \sim T_2$, coherence enhancement scales as $(T_2/\tau)^2$, yielding $\tau_{100} \sim 0.05 T_2$. On the other hand, coherence decay of nuclear spin qubits is sensitive to fluctuators in regimes (a)–(c) with $\Gamma T_2 \lesssim 1$, which makes the threshold τ_c somewhat smaller than T_2[26]. Here $\tau_{100} \sim 0.03 T_2$, showing a smooth crossover from $1/I$ to $1/I^3$. Our study shows that the application of π-rotations every $\tau \sim 0.1$ μs for an electron spin qubit and $\tau \sim 10$ μs for a nuclear spin qubit is sufficient to increase coherence by two orders of magnitude, effectively reaching the phonon emission limit[7].

IV. DISCUSSION AND CONCLUSION

Fig. 2 answers the previously open question of the dependence of spectral diffusion with increasing I. There had been earlier speculation[27] that the spectral diffusion rate $1/T_2$ might decrease with increasing I because of motional narrowing [due to faster flip-flops - notice that averaging over the I' subspaces in Eq. (15)results in $\Gamma \propto I^3$]. We show here (see Fig. 1) that the drastic increase in size of the Hilbert space for increasing I does actually compensate for the strong amplification of flip-flop rates and that therefore the system does not enter the motional narrowing regime, resulting instead in more spectral diffusion induced decoherence as I increases.

Another interesting point to note is that the repetition rate needed to suppress spectral diffusion (of the order of $I^2\times$MHz) is significantly different than that corresponding to the strength of dipole-dipole interactions, which are given by the root mean square of the last two terms of Eq. 1. This root mean square average is commonly referred to as the square root of the second moment (see, e.g., Eq. 3.53 of Ref. 15). For the systems of interest this is of the order of $I\times$KHz. The rate for controlling spectral diffusion is *significantly faster* than dipole-dipole coupling because hyperfine coupling (absent from the second moment equation) is very strong (up to MHz) and the spectral diffusion decay occurs due to the collective fluctuation of several nuclei under the field of the electron. We emphasize that the relative time scale for CPMG control of spectral diffusion and its dependence on the nuclear spin value I that we have established here can only be derived by carefully considering the combined effect of hyperfine interaction and collective dipolar fluctuation of 10^4 nuclear spins under the inhomogeneous hyperfine field produced by the electron, as we have done in this paper.

The effectiveness of the CPMG sequence in controlling spectral diffusion depends largely on the availability of fast methods for precise spin manipulation. The pulsing time has to be much smaller than τ, while the spin rotation angle should be precisely tuned to π[28]. The train of π pulses must also be applied without heating the sample above the required ~ 100 mK temperature. Currently, there are several proposals for overcoming this technical challenge, including all electrical spin resonance[18], optical manipulation[19], and implanting a low power microwave source nearby the qubit[29]. If this technical problem can be solved, our work indicates that the CPMG sequence will play a crucial role in extending electron spin coherence times in semiconductors.

In conclusion, we have provided a scheme for coherence control of localized spins in semiconductors subject to general nuclear spin-I fluctuations. We find that the rate for π-pulsing needed for substantial coherence enhancement is $\sim I^2$, yielding an efficient route for achieving long time spin coherence in semiconductors using advanced pulse technology.

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